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Simulation of Organic Chemical Movement in Hawaii Soils with PRZM: 2. Predicting Deep Penetration of DBCP, EDB, and TCP[1]

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ABSTRACT: PRZM was employed to simulate deep leaching of three fumigant chemicals beneath a central Oahu pineapple field. Our results suggest that PRZM, although not deployed here within the range of conditions for which the model was developed, can be a useful tool for making pesticide leaching assessments in Hawaii.

AN ENVIRONMENTAL ISSUE of national concern is nonpoint-source pollution of key regional groundwater systems by organic chemicals. A major cause of this kind of contamination is leaching of agricultural chemicals in aquifer recharge areas. This type of problem is magnified for insular systems, like those in Hawaii, because an alternative potable water supply is usually not readily available.

The freshwater lens of an oceanic island is a precious commodity and requires protection. It is not always possible to foresee what activities will harm such a system. For example, trace amounts of 1,2-dibromo-3-chloropropane (DBCP), ethylene dibromide (EDB), and 1,2,3-trichloropropane (TCP) were recently discovered within the Pearl Harbor aquifer on the island of Oahu (Oki and Giambelluca 1987). Each of the detected chemicals can be linked to soil fumigants employed by pineapple growers to control nematode populations; both DBCP and EDB are volatile fumigants while TCP is an impurity in a third fumigant known as DD (a mixture of 1,3-dichloropropene and 1,2-dichloropropane).

For more than 30 years it was generally believed that pesticides used by the pineapple industry in Hawaii would not leach beyond the near-surface zone. This conclusion was based on the high volatility and sorption of fumigants in the surface soils. However, measured concentrations of DBCP, EDB, and TCP down to 30 m at several locations proved the original assessment wrong and resulted in an urgency to know if the replacement chemicals used today will also leach to significant depths. Mathematical models with appropriate input data for local situations constitute a rational approach for assessing chemical leaching under a wide range of soil and climatic conditions.

This paper is the second part of a two-part series in which an institutional model, known as the Pesticide Root Zone Model (PRZM), was tested for the special case of pesticide leaching in structured Hawaii soils under pineapple culture. In the first paper (Loague et al. 1989), we reported on the ability of PRZM to predict observed concentration profiles for EDB. The simulations reported here differ from those in the first paper in several ways: (1) a superior scheme was used to disaggregate rainfall from monthly to daily values; (2) degradation was considered for some EDB simulations; (3) supplemental hydrogeologic data were used for some EDB simulations; and (4)

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concentration profiles were simulated for DBCP and TCP.

PRZM (Carsel et al. 1984) was developed by an interdisciplinary group of EPA scientists to simulate one-dimensional transport of a single pesticide within, and at shallow depths below, the unsaturated root zone. The two major components of PRZM, utilized in this study, are a water-balance algorithm and a chemical-transport algorithm. The water-balance algorithm is made up of three simple equations that partition water within and between the surface, the active root zone, and the remainder of the unsaturated zone. The elements of the water balance include precipitation, interception, evapotranspiration, runoff, and recharge. The water-balance calculations are performed on a daily time step.

The chemical-transport algorithm of PRZM is an implicit finite-difference approximation to the one-dimensional advection-dispersion equation. Solution of the transport equation necessitates soil water content and velocity values throughout the soil profile at each time step. These values are passed to the chemical-transport algorithm from the water-balance algorithm. The form of the advection-dispersion equation employed by PRZM in this study also includes the effects of sorption and degradation.

PRZM was not designed to simulate pesticide movement over the extended depths included in this study. The structure of PRZM is best suited to areas that are dominated by deep, well-drained sands where the water table is near the surface. These conditions are not met for the pineapple fields in Hawaii. With this limitation in mind, we have applied PRZM beyond its intended range to determine if the model can provide reasonable estimates of pesticide mobility over multiyear periods. A brief review of PRZM, as applied to this study, is given by Loague et al. (1989). A complete description of the model is outlined by Carsel et al. (1984).

MATERIALS AND METHODS

Pineapple field 4201 is located within the Pearl Harbor watershed near Mililani (Figure 1).

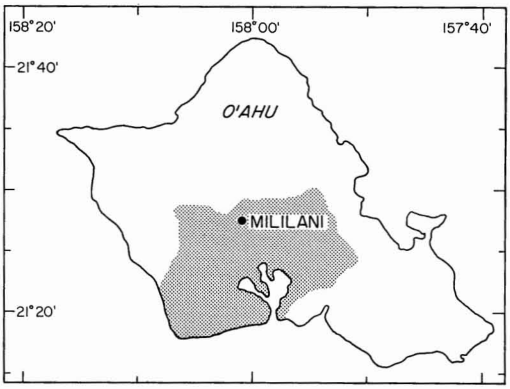


FIGURE 1. Location of the Pearl Harbor aquifer on Oahu (indicated by stippled area).

The cross-sectional geometry for a typical set of pineapple beds from 4201 is illustrated in Figure 2. Fumigation dates and rates for 4201 are listed in Table 1. Observed concentration profiles of DBCP, EDB, and TCP for a particular 4201 site known as 4201a are shown in Figure 3 for 1983 and 1985. Near-surface soil organic carbon data were collected for this study to supplement the information used in the first part of the investigation (Loague et al. 1989). Figure 4 shows average organic-carbon values for nine auger holes surrounding 4201a. The organic-carbon distribution coefficient ($m^3\ kg^{-1}$) values used for DBCP, EDB, and TCP in this study are 3.0×10^{-2}, 5.7×10^{-3}, and 7.8×10^{-3}, respectively. The DBCP value was measured by Williams et al. (1988) for Hawaiian conditions. The EDB and TCP values were estimated from the ratio of the octanol-water content and organic-carbon distribution coefficients for DBCP and the EDB and TCP octanol-water content coefficients.

Cases

We used PRZM to simulate pesticide mobility at 4201a for six separate cases: A, base case; D, volatilization; E, degradation; F, volatilization and degradation; G, detailed characterization of soil/saprolite profiles (Miller 1987); and H, new base case. Table 2 summarizes the experiments reported in this study. Case A includes sorption, dispersion,

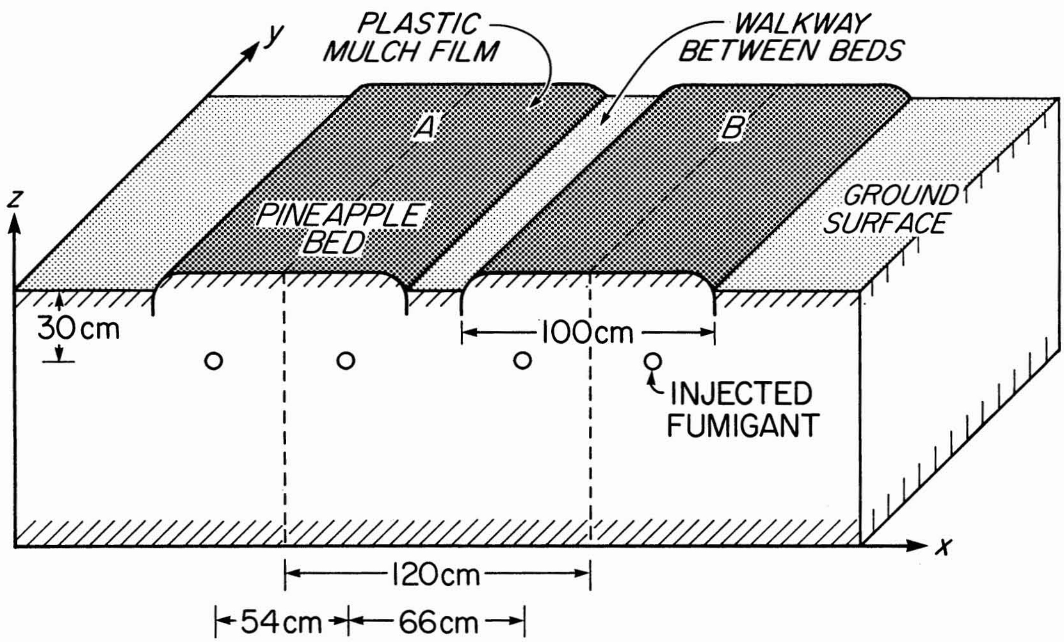


FIGURE 2. Cross-sectional geometry for two pineapple beds.

TABLE 1

FUMIGATION DATES AND APPLICATION RATES FOR FIELD 4201

APPLICATION DATE	APPLICATION RATE (kg/ha)		
	DBCP	EDB	TCP[a]
25 May 1950	—	—	22.51
2 September 1953	—	—	26.83
5 April 1957	—	—	26.96
2 June 1960	71.16	—	27.94
10 July 1964	78.93	—	30.05
16 June 1968	104.99	—	29.09
1 July 1972	112.76	—	26.12
28 September 1976	138.04	—	31.99
1 September 1981	—	241.37	—

[a] A 5% active ingredient of TCP is assumed for DD (Carter 1954).

and unrestricted drainage. Cases B and C, reported by Loague et al. (1989), considered the effects of setting the dispersion coefficient to zero (case B) and restricted drainage (case C). Cases D–F are modifications to the base case. The near-surface hydrologic and hydrogeologic characteristics of 4201a used to excite PRZM for cases A and D are given in Tables 3 and 5 of Loague et al. (1989).

Cases E–H are new to this study. Case E is identical to case A except that degradation is considered. The pesticide decay rate coefficient for EDB is taken as 0.005 $days^{-1}$, uniformly with depth, for this study. This value was determined by integrating under both the 1983 and 1985 EDB concentration profiles for 4201a and assuming that the difference in mass was due solely to degradation. Case F is the same as case D except that degradation is also included. Case G utilizes supplemental hydrogeologic information reported by Miller (1987) that was not available when the original analyses were conducted. Case G parameter values different from those in case A are listed in Table 3. Case H is described in the discussion section.

Volatilization

Each of the three fumigants of concern in this study has a relatively high Henry's law constant and therefore they are likely to evapo-

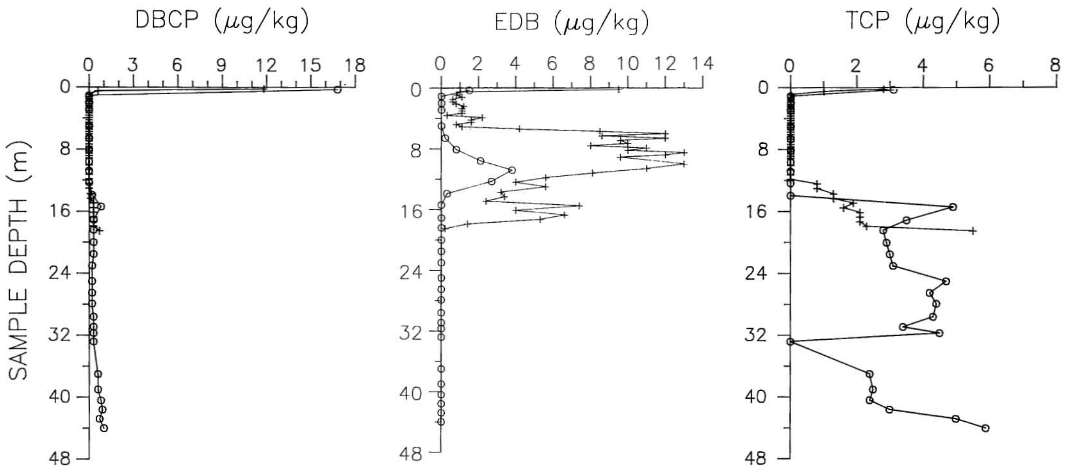


FIGURE 3. Observed DBCP, EDB, and TCP concentration profiles for 4201a (after Oki and Giambelluca 1985). The + and O represent 1983 and 1985 measurements, respectively. The 1983 and 1985 measurements were made, respectively, by the Hawaii State Department of Agriculture and the Water Resources Research Center at the University of Hawaii.

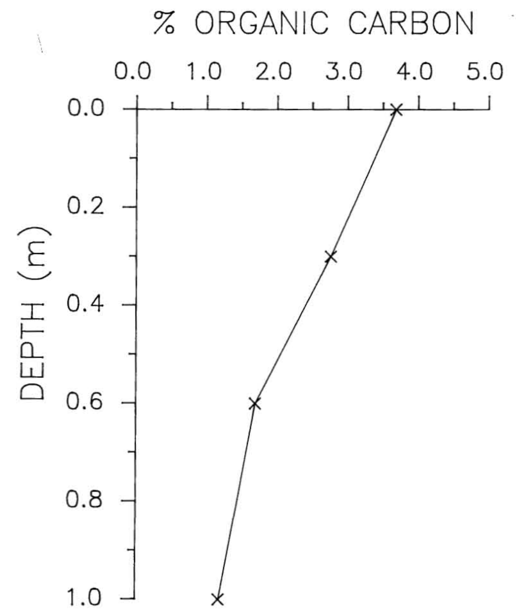


FIGURE 4. Near-surface organic-carbon profile for 4201a.

TABLE 2

SUMMARY OF PRZM SIMULATIONS

	CASE[a]					
CHEMICAL	A	D	E	F	G	H
DBCP		X				
EDB	X	X	X	X	X	X
TCP		X				

[a] A, base case; D, volatilization; E, degradation; F, volatilization and degradation; G, detailed characterization of soil/saprolite profiles; H, new base case.

rate to the atmosphere. PRZM, however, has no vapor-phase component and subsequently cannot account for volatilization. Because PRZM is not well suited for volatile chemicals we have elected here to estimate short-term volatilization effects with a separate model. The vapor-phase model developed by Green et al. (1986) was selected to simulate residual pesticide concentrations. The results from the preprocessing were in turn used as input for PRZM to more realistically approximate initial conditions for the volatile chemicals.

Application of the three pesticides at 4201 was done by chisel injection under plastic mulch (see Figure 2). Immediately after injection, the fumigants are in the form of a line source. These systems are symmetrical; therefore, the two-dimensional model of Green et al. (1986) is sufficient to simulate initial mixing of residues in the top soil cross-sectional plane.

TABLE 3

Soil Parameters for 4201a (Adapted from Miller 1987)

DEPTH (m)	BULK DENSITY ($kg\ m^{-3}$)	SOIL WATER–FIELD CAPACITY (dimensionless, assumed at 33 kPa)	SOIL WATER–WILTING POINT (dimensionless, assumed at 1.5 MPa)
0.0– 2.0	1,140	0.53	0.45
2.0– 3.5	1,020	0.60	0.51
3.5– 5.5	1,210	0.49	0.35
5.5– 7.0	1,210	0.50	0.37
7.0– 8.5	850	0.63	0.43
8.5–11.0	980	0.57	0.43
11.0–13.0	1,090	0.54	0.44
13.0–15.0	990	0.62	0.38
15.0–20.0	990	0.57	0.40
0.5–20.0	1,270[a]	0.42[a]	0.25[a]

[a] Values used for case A (Loague et al. 1989).

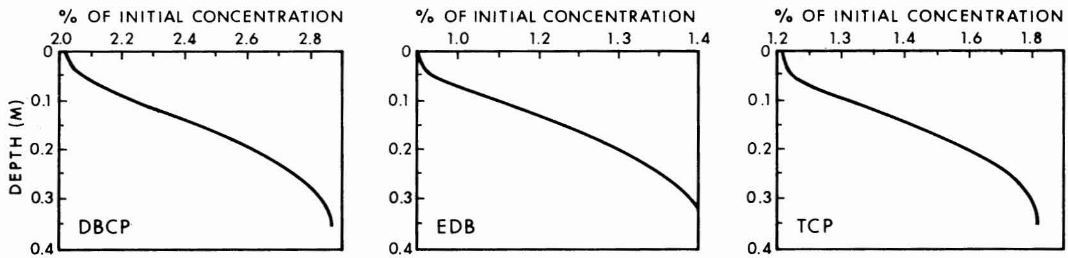


Figure 5. Distribution of residues for (*left*) DBCP, (*middle*) EDB, and (*right*) TCP 14 days after application.

Recharge under pineapple crops is usually minimal when fumigants are applied. Therefore, initial mixing is accomplished by the action of vapor-phase diffusion. The total amount of residue in the soil is partitioned between the vapor, liquid, and solid (sorbed) phases. The lateral spreading and loss by volatilization of pesticides depend, in general, on the sorption coefficient and the Henry's law constant.

The effective diffusion coefficient in the governing equation of the two-dimensional model (Green et al. 1986, Loague et al. 1989) is a function of chemical (sorption coefficient, Henry's law constant) and soil (soil water, air-filled porosity, bulk density) parameters. Among these parameters, the sorption coefficient is the most important in determining initial mixing and volatilization (Green et al. 1986). For the present study the sorption coefficients ($m^3\ kg^{-1}$) for DBCP, EDB, and TCP for 4201a were estimated to be 1.2×10^{-3}, 2.2×10^{-4}, and 3.0×10^{-4}, respectively, using the organic-carbon distribution coefficients for the three pesticides and the organic-carbon contents shown in Figure 4. The values of the other parameters are the same as given by Loague et al. (1989).

Figure 5 shows the distribution of residues for DBCP, EDB, and TCP in the top 0.4 m of soil at the end of 2 weeks after fumigant application. The average concentration of residues calculated from these distributions was used as initial values for the PRZM long-term simulations. A time of 2 weeks was used for initial mixing because a reasonably uniform lateral distribution of fumigant residues was achieved and the further reduction of residue concentration after this time became rather small.

Rainfall

Daily rainfall inputs for PRZM were estimated from monthly data. The monthly rainfall data were estimated from nearby rain gauge data. Three schemes were used to convert monthly information into daily data for various simulations: I, monthly totals applied to the first day of the month; II, monthly totals spread uniformly over each day of the month; and III, monthly totals disaggregated to daily with a first-order Markov chain model. Pan evaporation data were also disaggregated for this study. Monthly evaporation totals were either applied to the first day of the month or spread uniformly over each day of the month. The former approach was used with rainfall scheme I while the latter approach was used with rainfall schemes II and III.

The rainfall disaggregation model used in scheme III was developed by Giambelluca and Oki (1987). To simulate daily rainfall and at the same time utilize the information provided by the estimated monthly rainfall totals, a nondimensionalized parameter Y is used:

$$Y = P_d P_m^{-1}$$

where P_d is daily rainfall (mm day^{-1}) and P_m is monthly rainfall (mm day^{-1}). The chain of dimensionless Y values can subsequently be multiplied by the monthly rainfall to obtain a simulated daily rainfall chain. The probability density functions and transitional probability matrices, the primary components of the Markov chain model, were estimated from a sample of daily rainfall at a nearby station. The Weibull distribution was found to adequately fit the sample data and was used in the simulation. The continuous Weibull distribution was divided into nine discrete non-zero rainfall ranges for each of two seasons. Values of Y were simulated with the use of a random number generator. Daily rainfall depths were determined as the product of Y and the monthly rainfall.

The scheme III disaggregation model enabled us to produce stochastic daily rainfall sequences each of which sum to the estimated monthly rainfall total. For a given month, any number of realizations of daily rainfall sequences could be generated. Five daily rainfall simulations were computed for each month to examine the effects of different realizations on solute transport predictions. Although the simulated daily rainfall possesses the same frequency distribution and autocorrelation as the historical rainfall of the region, the model cannot reproduce the actual sequence of events during any given month. The advantage of this approach is that in the absence of measured daily rainfall, likely sequences of daily rainfall can be generated from monthly rainfall. The model makes use of all available information including regional daily rainfall characteristics and known monthly rainfall totals.

The five rainfall time series realizations were employed separately as input for PRZM simulations. The five resulting concentration profiles were averaged for a final concentration profile. An example of component and averaged concentration profiles is shown in Figure 6. Rainfall scheme III was the primary procedure used in this study to glean daily data from monthly data. The disaggregation approach is not suggested as a replacement for daily measurements. We used scheme III to estimate information that can never be truly known unless it is measured.

RESULTS

The PRZM simulations reported here were each for a depth of 20 m. The space increment throughout the profile for this study was 0.1 m. The decision to limit predictions with PRZM to the 20-m depth was based on the characteristics of the observed concentration profiles, computer costs, and the large number of simulations attempted. We feel that the 20-m depth is more than sufficient to characterize the general nature of the model performance. The observed and predicted concentration profiles reported here, for various years, are all for the month of September. Much of the effort in this study was placed on EDB because there was only a single application of the chemical and because concentration profiles were measured on two dates after the application. Both DBCP and TCP (DD) were applied several times before the concentration profiles

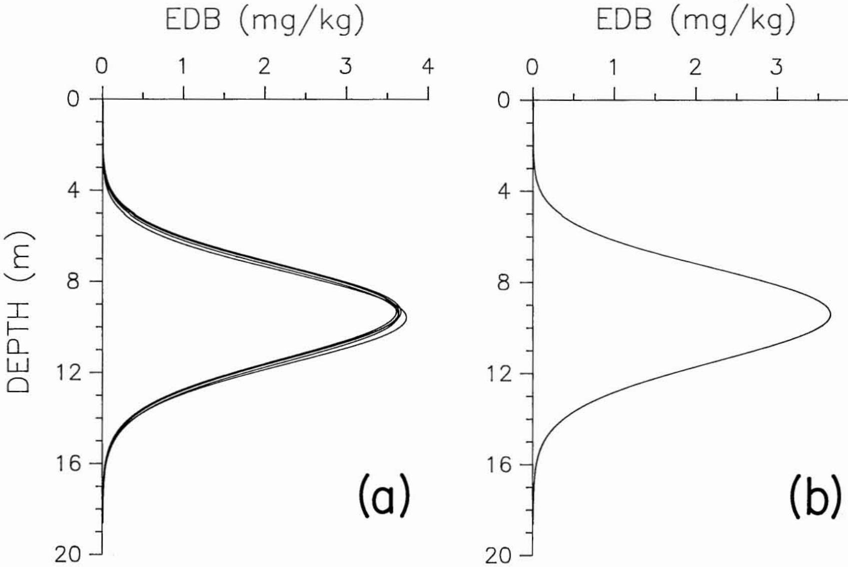


FIGURE 6. (*a*) Five EDB concentration profiles resulting from five separate rainfall scheme III realizations. (*b*) Average EDB concentration profile resulting from component EDB concentration profiles in (*a*).

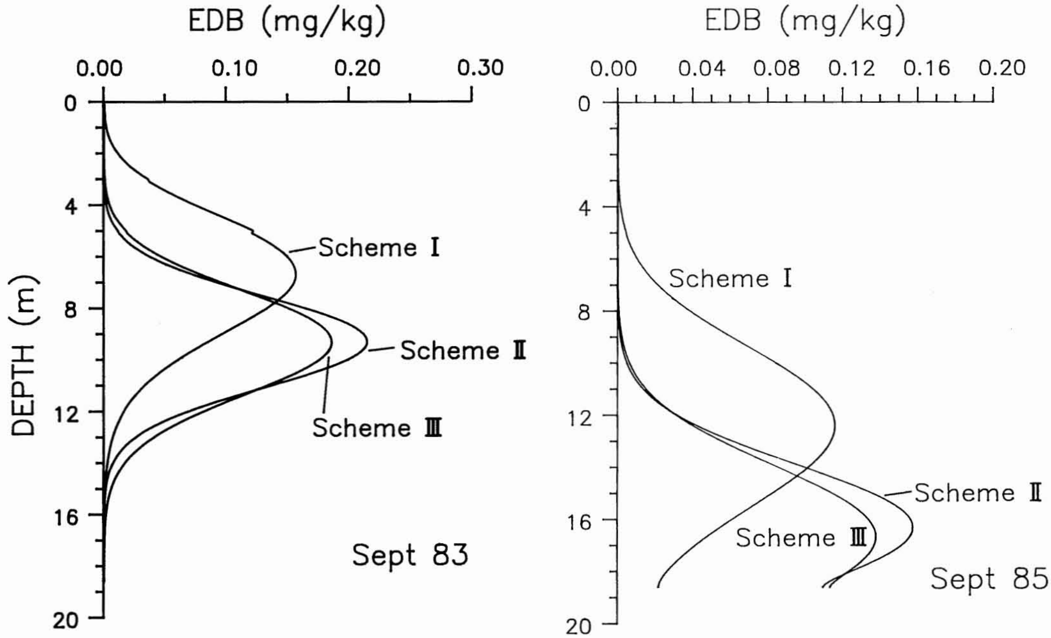


FIGURE 7. Predicted EDB concentration profiles for 4201a for case A for rainfall schemes I, II, and III for 1983 and 1985.

were measured, making the interpretations for these chemicals more difficult than those for EDB.

Figure 7 illustrates the consequence of each of the three rainfall schemes for predicted EDB concentration profiles. Figure 8 presents once-a-year predicted EDB concentration profiles for cases A and D–F for a 4-yr period starting

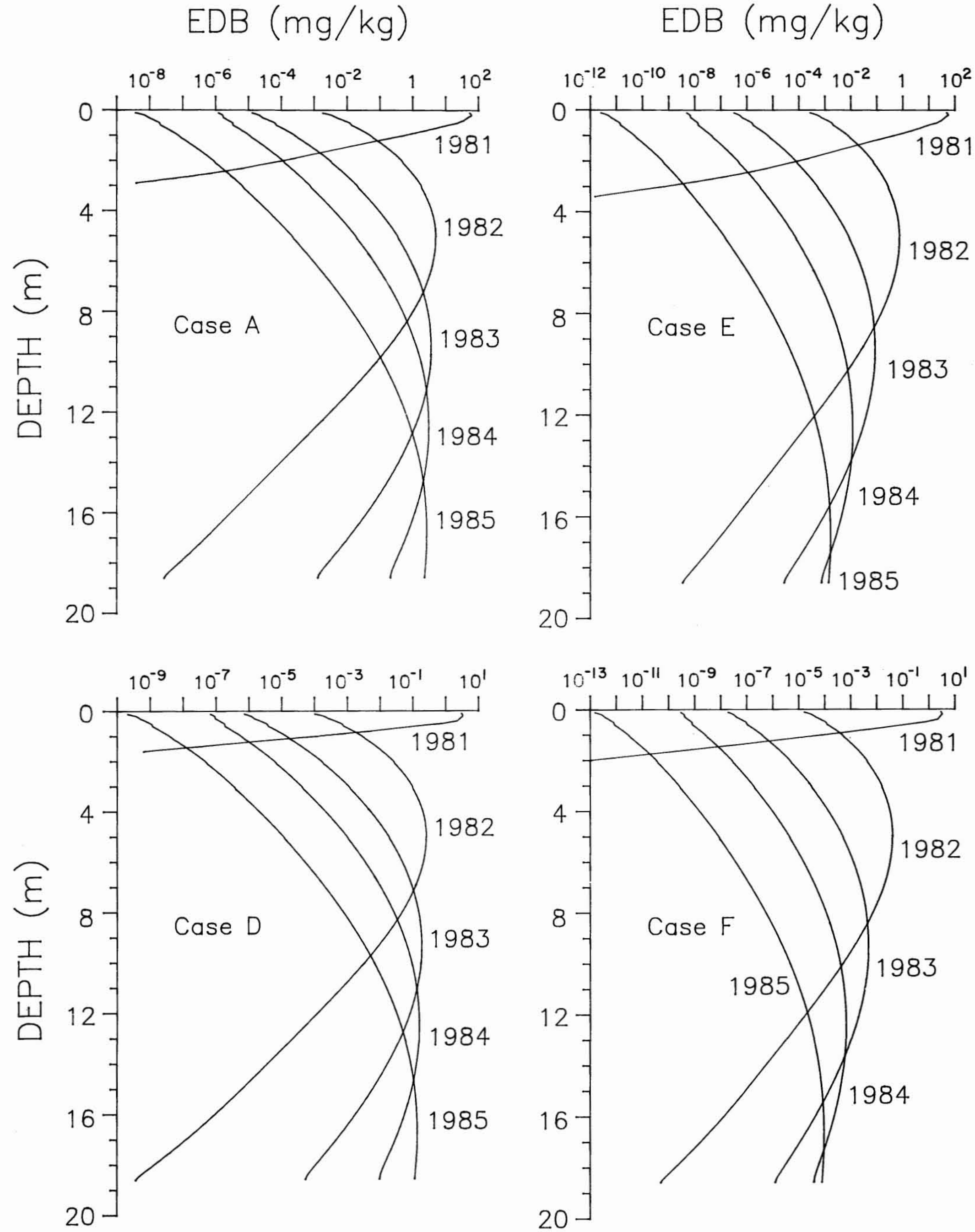


FIGURE 8. Predicted EDB concentration profiles for 4201a for case A, D, E, and F for rainfall scheme III for 1981–1985. Note that each graph has a different range of concentrations.

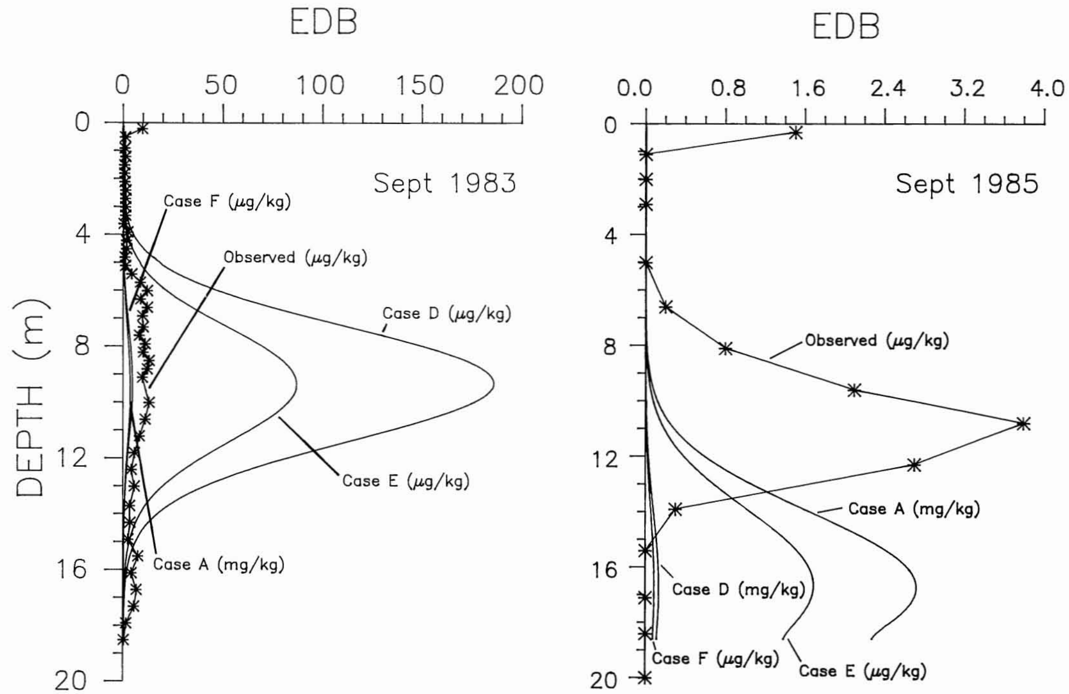


FIGURE 9. Composite overlays of predicted EDB concentration profiles for 4201a for cases A, D, E, and F for rainfall scheme III for 1983 and 1985 versus observed data. Note that different units are used for the various curves.

just after the application date. Figure 9 shows composite overlays of predicted and observed EDB concentration profiles. Figure 10 summarizes predicted concentrations of DBCP and TCP at 10 m for case D. Figures 11 and 12 present one-a-year predicted DBCP and TCP concentration profiles, respectively, for case D. Figure 13 shows predicted and observed DBCP and TCP concentration profiles for case D. Figure 14 shows an overlay of predicted EDB concentration profiles for cases A and G.

Inspection of Figures 7–14 leads to the following comments:

1. Simulated chemical leaching is at a slower rate for scheme I than for schemes II and III (see Figure 7). The depth of the maximum concentration peak at a given time is similar for schemes II and III. The concentration peak for scheme III is between that of schemes I and II. Scheme I results in lower predicted recharge (Loague et al. 1989) whereas schemes II and III more closely represent perceived recharge conditions. We feel that scheme III is the most realistic of those tested and that scheme II is generally superior to scheme I. Scheme I is similar to a monthly water balance model, which would tend to underestimate recharge.

2. The combined effects of initial short-term volatilization and long-term degradation are considerable when compared with simulations that do not account for these processes (see Figure 8). For example, EDB concentration peaks are reduced by several orders of magnitude from case A to case F when both volatilization and degradation are considered. It appears, at least for EDB at 4201a, that accounting for degradation reduces the residual error more than preprocessing for volatilization (see Figure 9). The simulation scenario that best describes the 1983 observed concentration profile is the one that includes both volatilization and degradation (case F). For 1985, however, the better fit is from the simulation that includes only degradation

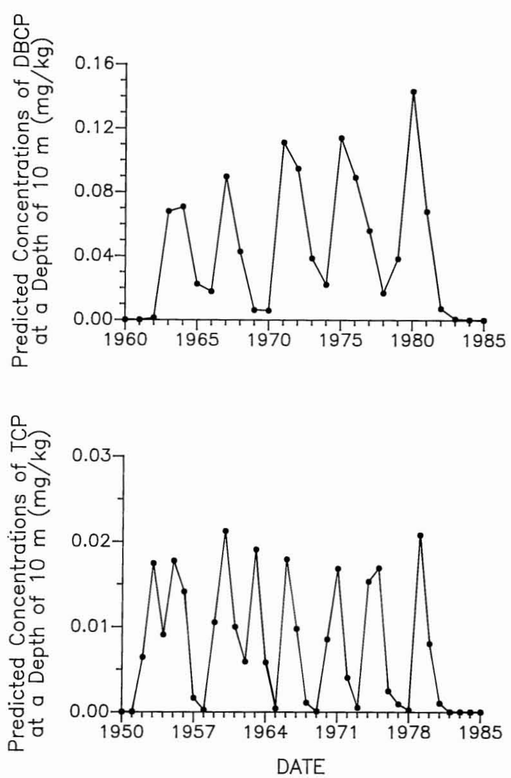


FIGURE 10. Predicted concentrations of DBCP and TCP for 4201a at 10 m for case D for rainfall scheme III.

(case E). The observed EDB concentrations were, respectively, lower in 1983 and higher in 1985, than the case E predictions for those years. This suggests that the decay rate coefficient is set too low initially and too high a few years postapplication. Alternatively, this result may be due to an increase in EDB sorption over time as was observed by Green et al. (1986) for DBCP. Degradation rates will of course change with time. The sparse data used here do not allow for consideration of time-dependent degradation or sorption.

3. The lags between application dates and concentration peaks at 10 m for DBCP and TCP (see Table 1 and Figure 10) are well correlated. At this depth it is easy to identify individual events. The center of mass for either DBCP and TCP concentration profiles in Figures 11 and 12, for a given application, is at approximately 10 m at the time of the next application of the same chemical. These simulations, very generally, suggest that the center of mass is leaching at a rate of approximately 2.5 m per year. Of course, the fronts and tails of these concentration profiles are moving faster and slower, respectively, than is the center of mass, thus spreading out the profiles and causing them to eventually overlap. The predictions reported here for multiple application simulations, which cover 25 and 35 years for DBCP and TCP respectively, are in general not bad. This conclusion is based solely upon comparisons of the observed and predicted concentration profiles shown in Figure 13 for the two chemicals on two separate dates.

4. The concentration profiles shown in Figure 14 for cases A and G are somewhat different. The data used for these two simulations are identical except for those identified in Table 3. The bulk density is lower for case G than for case A while the field capacity and the wilting point are higher for case G than for case A. The variability in the case G data does not appear to have any spatial structure. The case A simulation, when compared to the case G simulation, estimates a smaller peak concentration but a faster moving front. These differences are obviously important for any assessment of pesticide mobility.

DISCUSSION

The PRZM simulations reported here are laced with uncertainty. In general, there are three sources of error inherent to pesticide mobility estimates (Loague et al. 1988): (1) model error; (2) input error; and (3) parameter error. The intent of a modeling exercise such as this one should be to minimize or at least recognize these sources of uncertainty and to determine what the consequences may be. To investigate the impact of input and parameter errors for the PRZM simulations reported here, several sensitivity experiments were performed. Figure 15 shows the results for EDB simulations with plus and minus 10% errors in: (a) daily rainfall values, (b) the distribution coefficient, (c) volatilization rates, (d) the decay-rate coefficient, (e) the hydrodynamic dispersion coefficient, and (f) the runoff curve

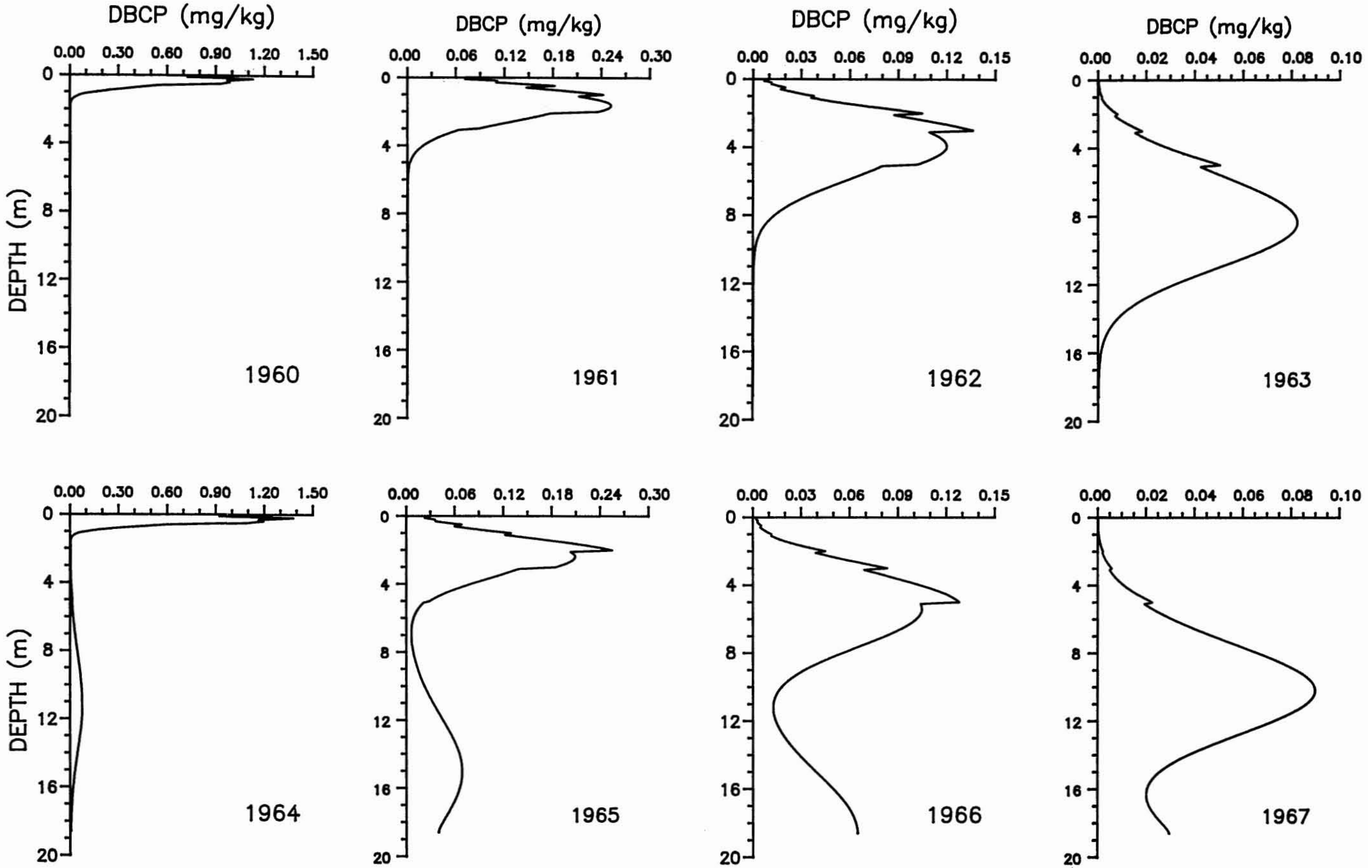
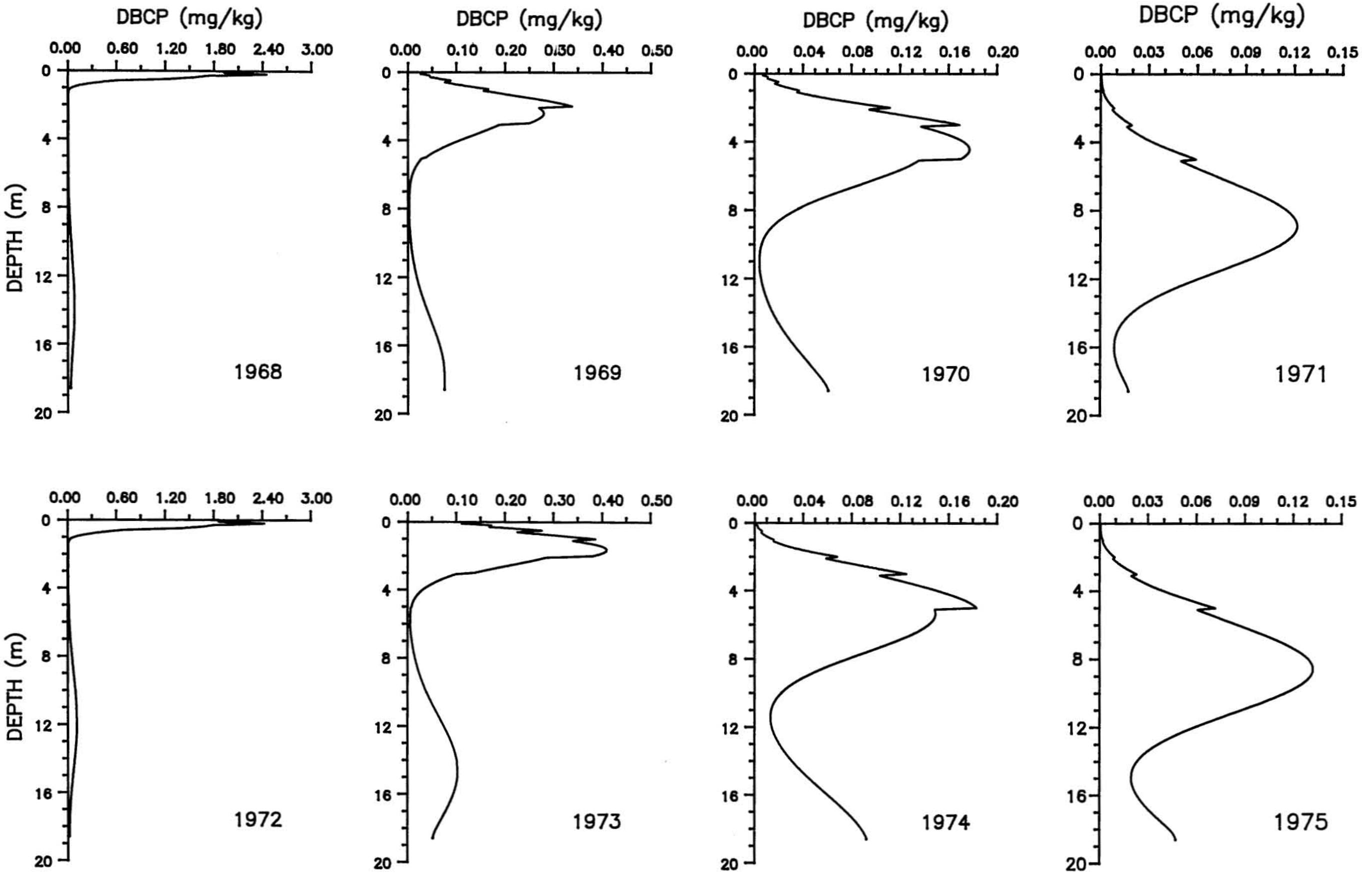


FIGURE 11. Predicted DBCP concentration profiles for 4201a for case D for rainfall scheme III for 1960–1985.



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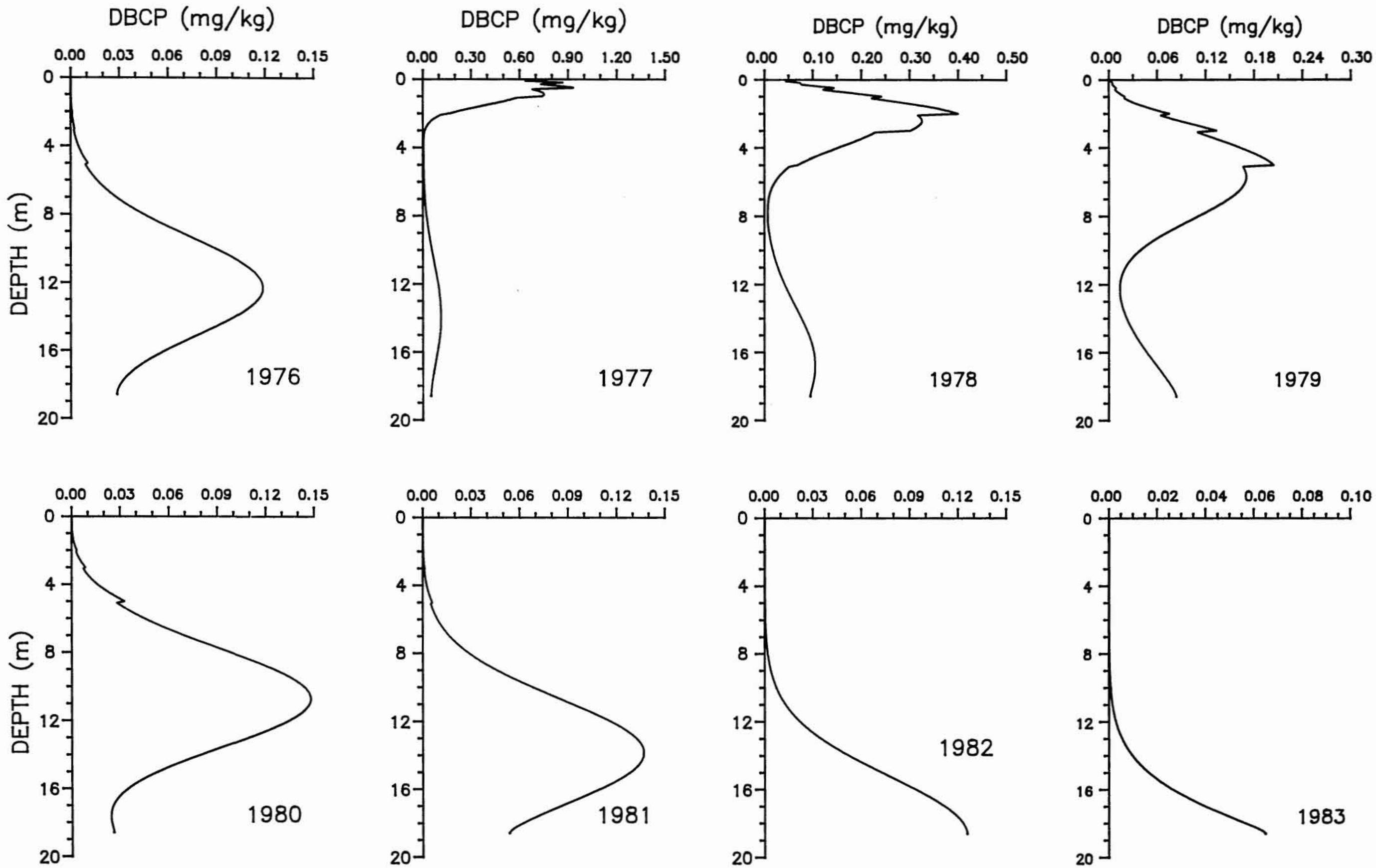

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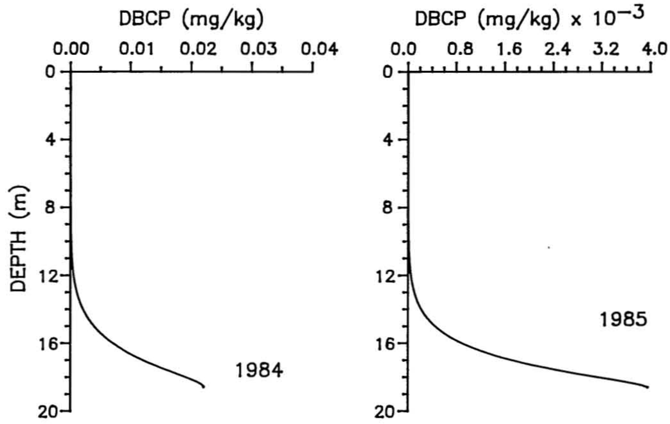


FIGURE 11 continued.

numbers. The standard for the simulations shown in Figure 15 is the new base case (case H). Case H is modified from case G by including both volatilization and degradation and is probably our best representation of 4201a with an uncalibrated PRZM. It is quite obvious that the largest impacts to the EDB concentration profile are caused by errors in the decay-rate coefficient and that errors in both the daily rainfall values and volatilization rates have some effect on predicted concentration peaks. The distribution coefficient and the runoff curve numbers are fairly forgiving for the range tested here. These results suggest that degradation, recharge, and volatilization are the most important processes to characterize for simulating EDB mobility at 4201a, and that sorption, dispersion, and runoff are less important for this particular case.

Degradation is not considered for the extended DBCP and TCP simulations with PRZM in this study. Decay-rate coefficients for these chemicals were not available. This could be considered a form of model error because a known process is being ignored. Degradation of EDB on the other hand was crudely estimated as a constant over a specific interval that was shorter than the total simulation period. The actual rate is almost certainly not a constant. This probably resulted in an overestimate of degradation when this rate was used for the entire simulation. Parameter error such as this will propagate even more as the simulation length is increased. Characterizing decay-rate coefficients for simulation of multiple pesticide applications, such as those for DBCP and TCP, could become quite complicated if concentration profiles were overlapping.

CONCLUSIONS

Mathematical models designed to simulate transient soil water movement and solute transport in unsaturated soil and hydrogeologic environments, subjected to nonuniform recharge rates, are timely tools for predicting pesticide mobility and can therefore be useful for asking "what if" questions and assessing groundwater contamination hazards. The primary limitation of deterministic-conceptual simulation for pesticide leaching is scarce data. The measured DBCP, EDB, and TCP concentrations for 4201a on two separate dates provide an excellent opportunity to investigate the predictive capabilities of PRZM. It was not our intention in this paper to rigorously calibrate and validate PRZM and then predict future migrations of DBCP, EDB, and TCP for 4201a. We have elected here, as in the first part of this study (Loague et al. 1989), to examine PRZM in an uncalibrated mode to highlight the characteristics of the model rather than perform yet another curve-fitting analysis.

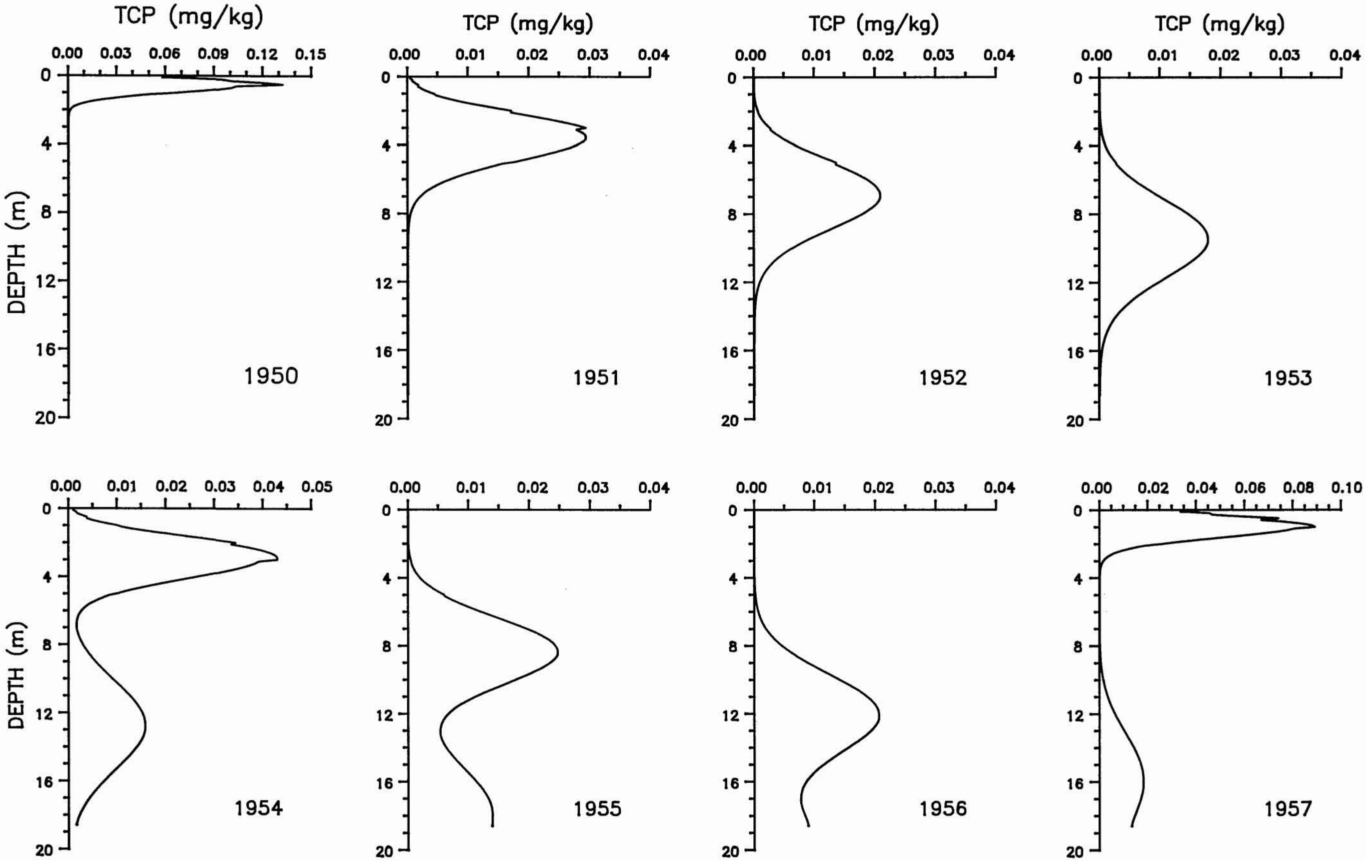
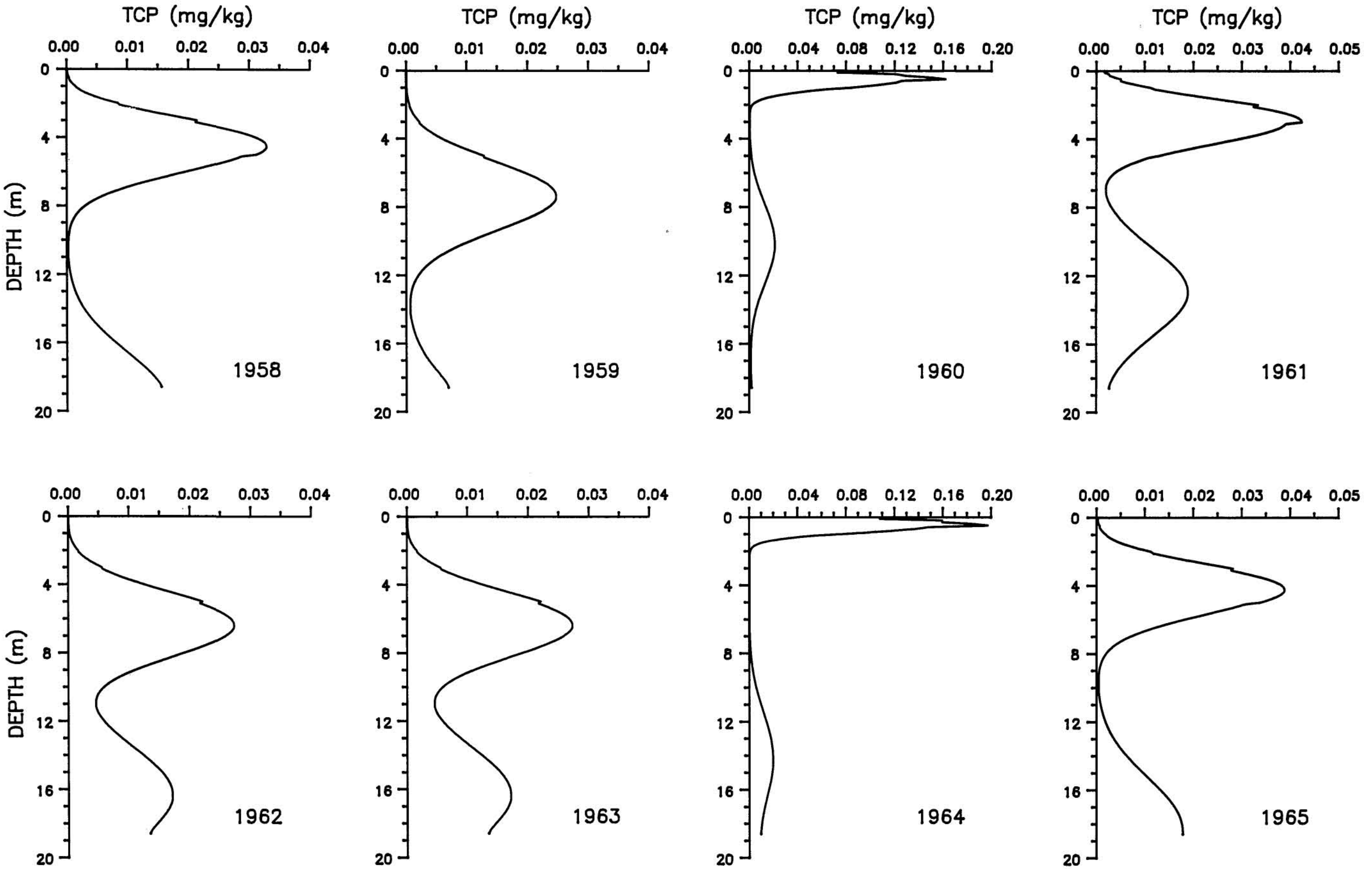


FIGURE 12. Predicted TCP concentration profiles for 4201a for case D for rainfall scheme III for 1950–1985.



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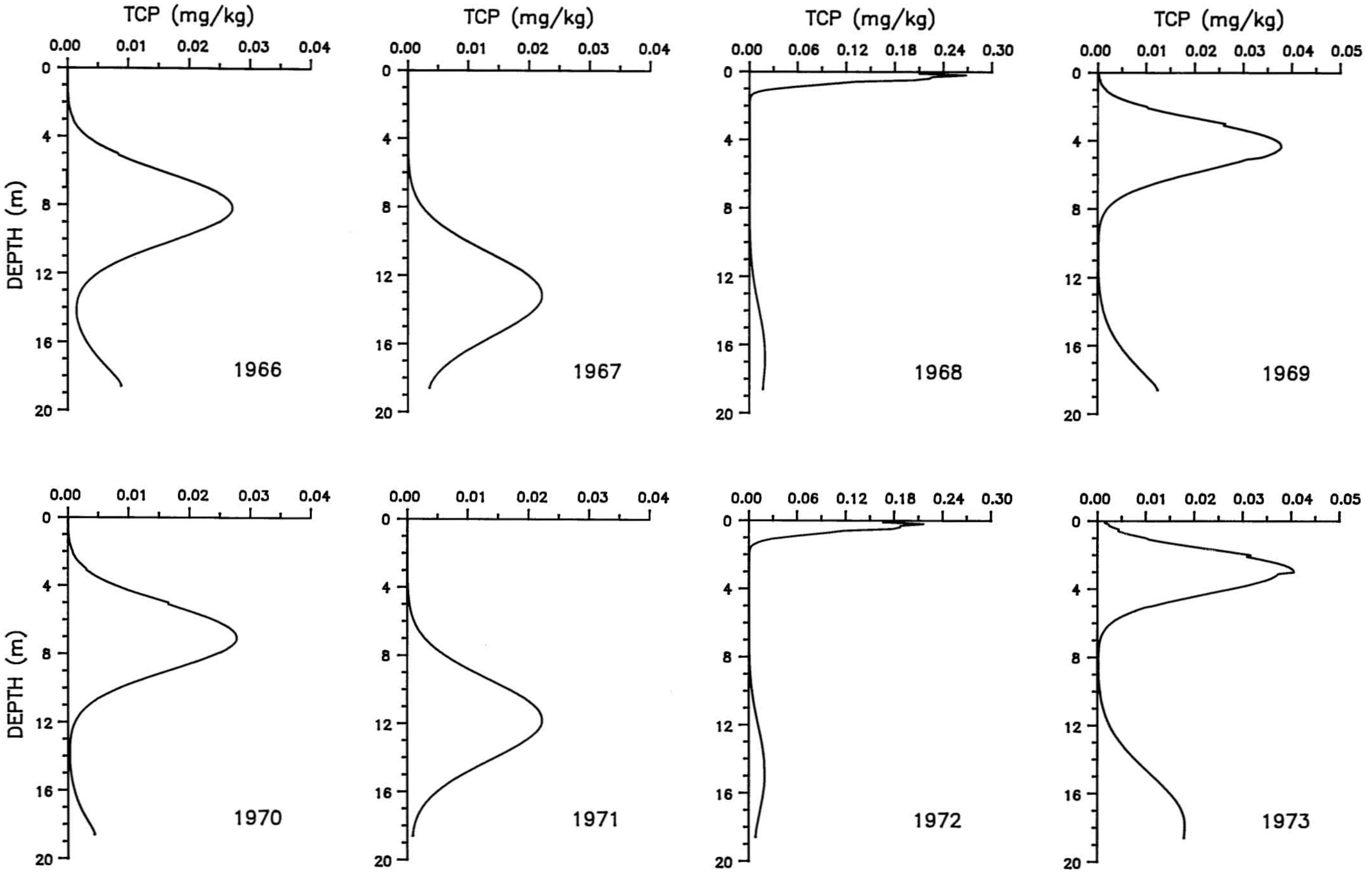
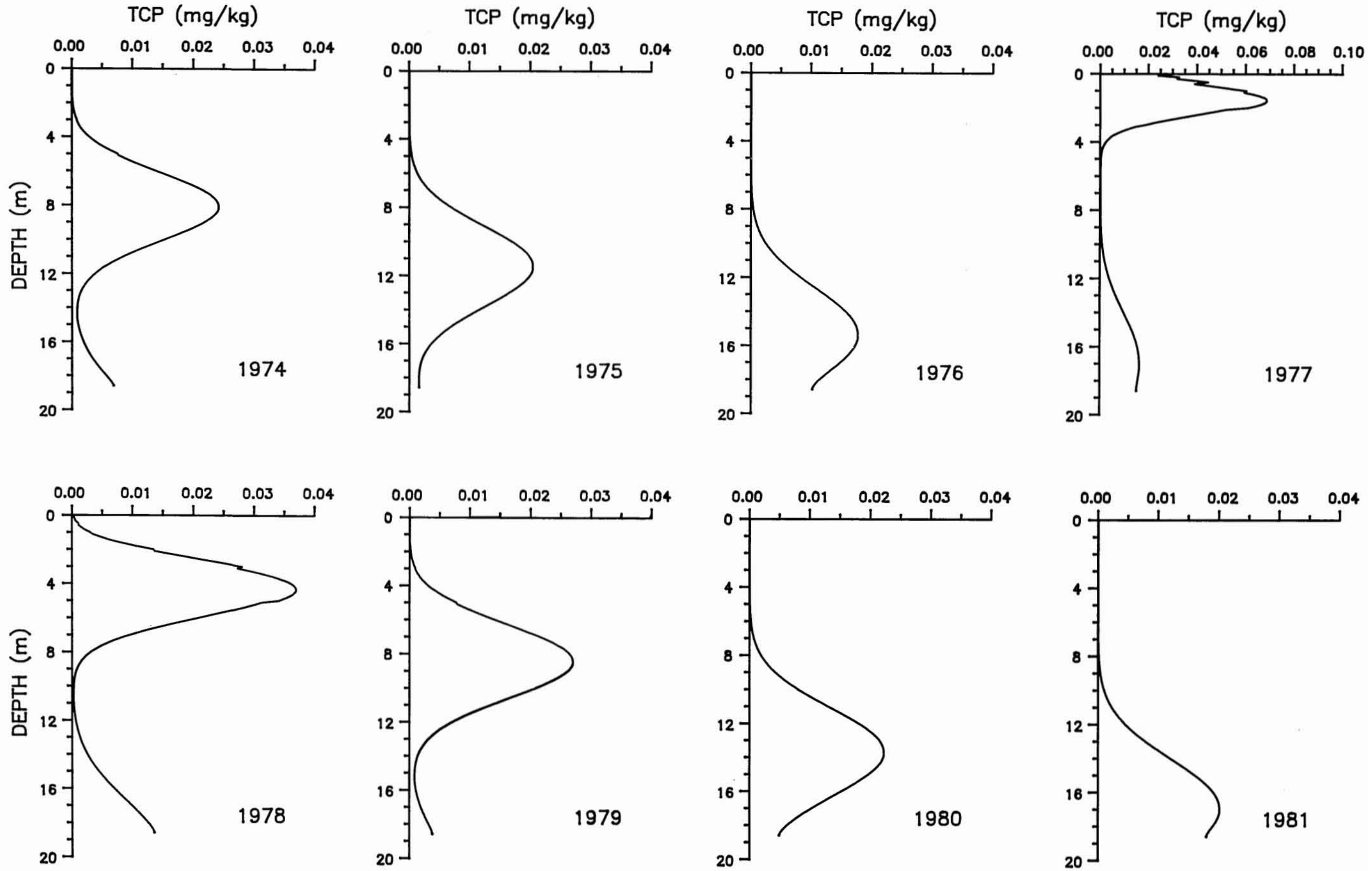


FIGURE 12 continued.



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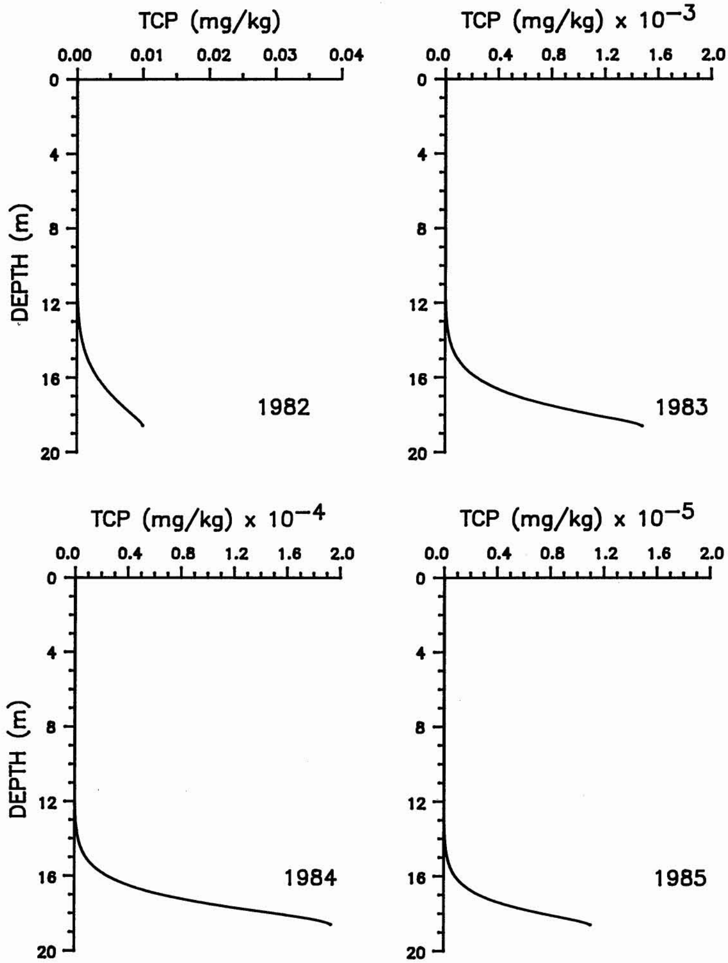

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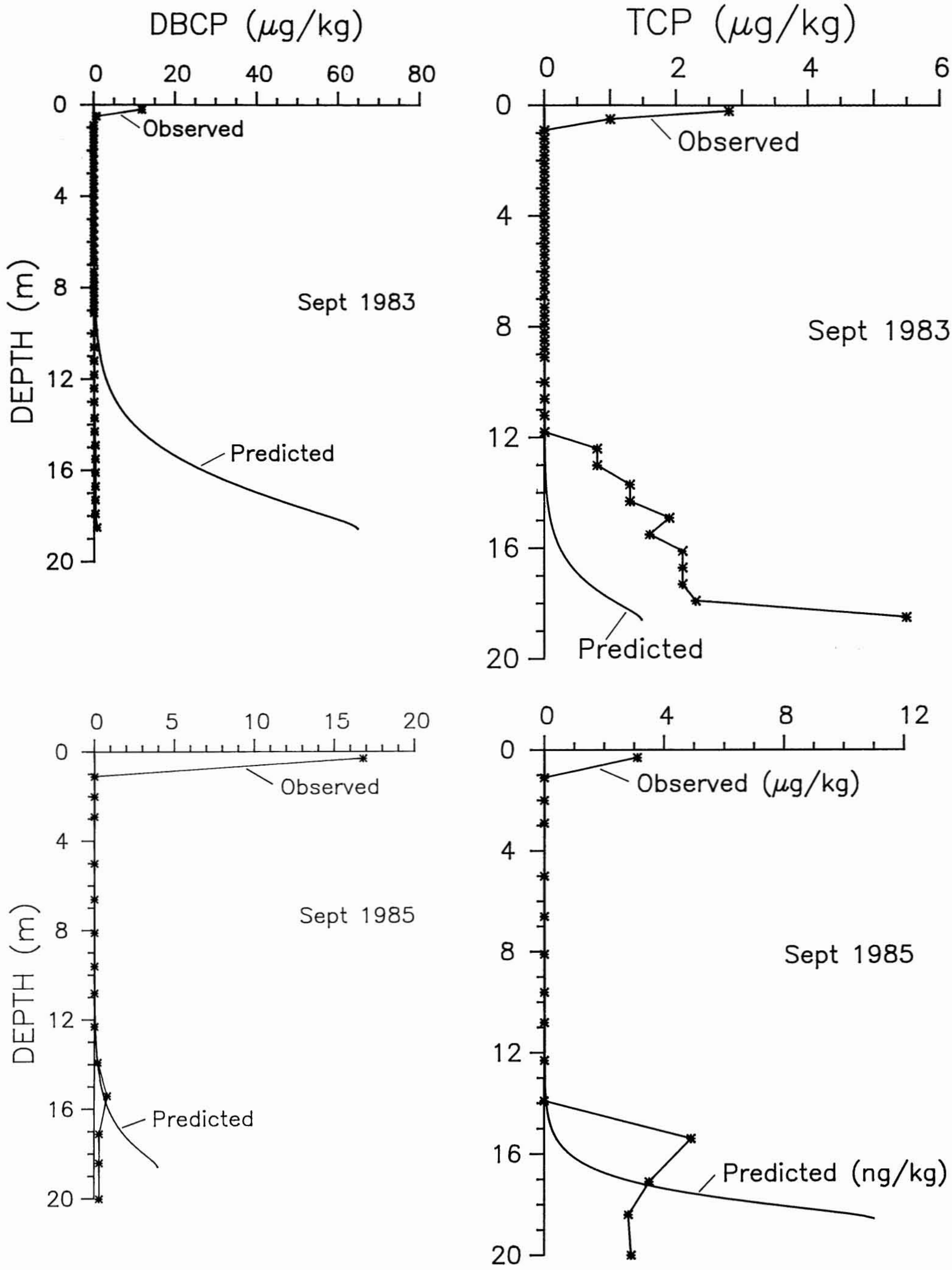

FIGURE 13. Predicted DBCP and TCP concentration profiles for 4201a for case D for rainfall scheme III for 1983 and 1985 versus observed data.

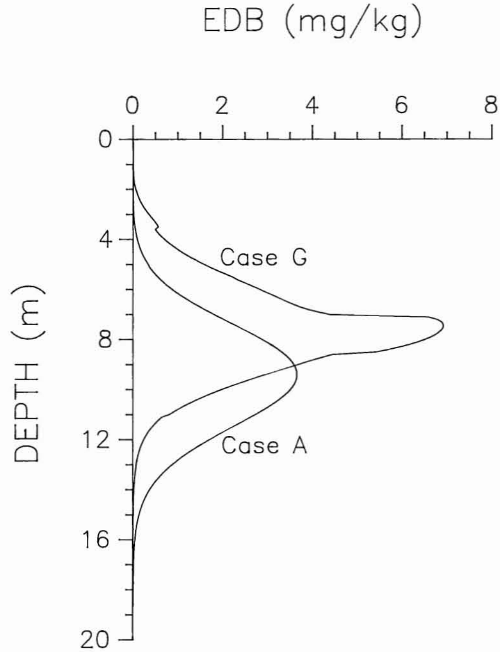

FIGURE 14. Comparison of predicted EDB concentration profiles for 4201a for cases A and G for rainfall scheme III for 1983.

In this paper we show that deep leaching of DBCP, EDB, and TCP is reasonably well predicted with PRZM at a single well-characterized site. We cannot, however, suggest that PRZM should be used in Hawaii for decision/management purposes until further testing has been conducted. The chemical and transport processes for volatilization, degradation, and preferential flow are not well defined. Significant amounts of a volatile chemical can be displaced by volatilization. PRZM has the potential for modeling the movement of volatile chemicals in soil, using a short-term preprocessing approach, but the model is far more appropriate for nonvolatile chemicals. Degradation kinetics must also be accurately simulated if PRZM-predicted results are to be realistic. Characterizations of decay-rate coefficients with depth are urgently needed for future work. Finally, fluid flow and solute transport through structured soil and fractured near-surface rock is an important, and as of yet unresolved, area of research, especially in Hawaii. PRZM is only concerned with a single mobile phase and does not allow for consideration of preferential flow. It may be possible to improve the performance of the model with some kind of mobile/immobile partitioning.

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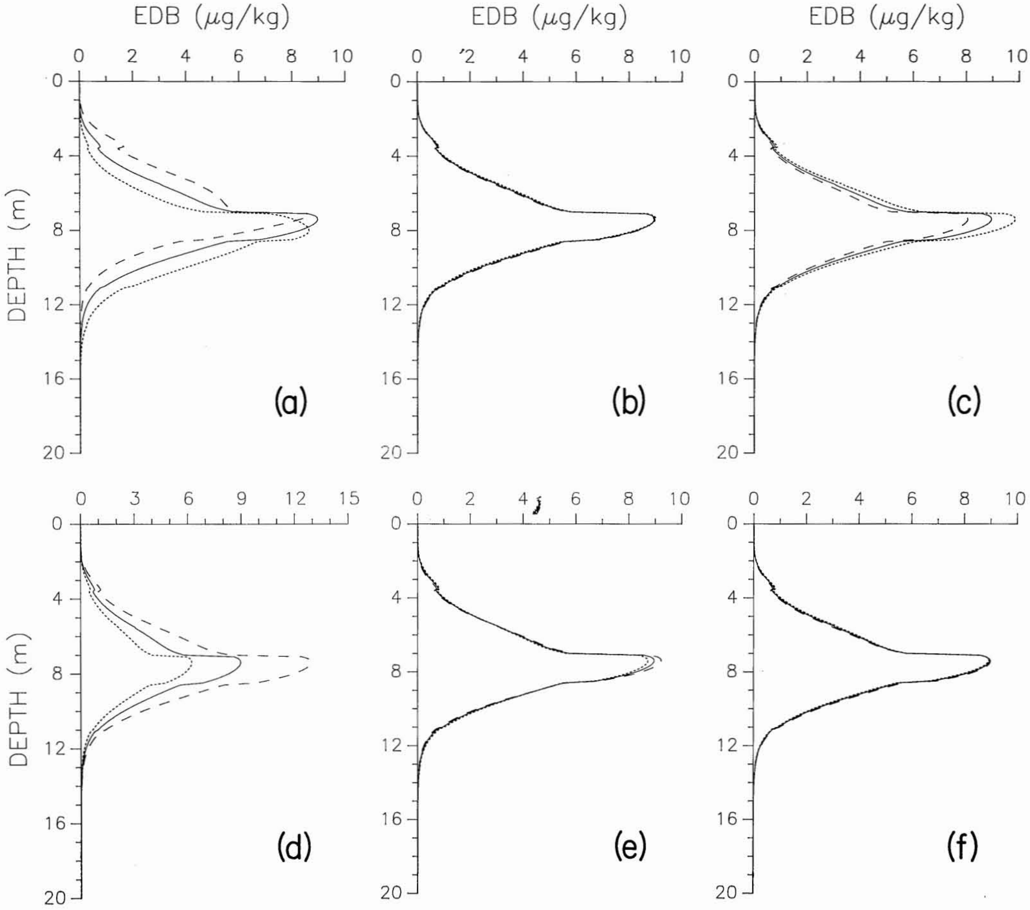

FIGURE 15. Sensitivity of predicted EDB concentration profiles for 4201a for case H for rainfall scheme III for 1983. Plus and minus 10% changes in: (a) rainfall rates; (b) the organic-carbon distribution coefficient; (c) volatilization rates; (d) the decay-rate coefficient; (e) the hydrodynamic dispersion coefficient; and (f) the curve number coefficients for runoff. The solid lines are case H, the dotted lines +10%, and the dashed lines −10%.

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